Holography of Acetylene Adsorption on Si(100)-2×1 Surface

S. H. Xu*, Y. Yang, E. Rotenberg[1], T. Yates, Jr.[2], and G. J. Lapeyre
Physics Department, Montana State University, Bozeman, MT 59717
[1]Advanced Light Source, LBNL, Berkeley, CA 94720
[2]Department of Chemistry, University of Pittsburgh, Pittsburgh, PA 15213

INTRODUCTION

An investigation of the adsorption of acetylene on Si(100) surface is important for the formation of silicon carbide, because the adsorption is the first step of whole reaction towards carbide formation. Previous studies[1,2] have proposed a di-σ model. In this model, the acetylene adsorbs on the bridge site across the Si dimer and the carbon-carbon triple bond is rehybridized to a di-σ bond state. However, there are some disagreements on the correct peak assignment in HREELS[1], and the exact nature of the initial bonding geometry of acetylene on the Si(100) surface is still a matter of discussion. In our work, the energy-dependent diffraction is obtained for C 1s core-level emission from the measurement of constant-initial-energy spectra(CIS) for a set of angles which forms a grid over the full-emission hemisphere. Then, we adopt the photoelectron holography to study the adsorption site and 3-D geometry of acetylene on the Si(100)-2×1.

EXPERIMENT

The experimental data is collected at the undulator beam-line 7 station at Advanced Light Source(ALS) in Berkeley. An angle-resolved electron energy analyzer was used to detect the C 1s photoelectrons. N-type Si(100) (4° vicinal cut) samples were prepared by outgassing at 700 K for ~20 hours, and then heated to 1180 K to yield the clean well 2×1 reconstruction. Research grade acetylene (99.9 mol % purity) was dosed when the sample was kept at room temperature(RT). After adsorption of 0.5 monolayer(ML) acetylene, there is no obvious change observed in LEED pattern. A set of 82 CIS was measured for the C 1s where the angles span an irreducible symmetry element, i.e., $0° \le \theta \le 75°$ and $0° \le \varphi \le 90°$. The azimuthal angles are referenced to a mirror plane along the Si dimer, $\varphi=0$. Both of azimuthal and polar angles are obtained by rotation of the sample. For each angle, C 1s signal is recorded as a function of the momentum(4.1–10.4 $Å^{-1}$) with a constant momentum $\Delta k=0.15$ $Å^{-1}$. Then we use the small-cone method[3] to get the image of this system.

RESULTS

The holographic image formed from C 1s is complicated, largely because two carbons(labeled as P and Q) in one acetylene molecule occupy inequivalent site. The transfer which converts data to an image put all emitters together at the origin. At the same time, each C atom is a scatterer for another one, then it can be "seen" by another C atom. From the x-y planar cut passing through the C emitters, we find that there are two spots P/Q and Q/P. The distance between them is 2.2 Å. However, the separation between the P/Q and Q/P spots is twice the C-C bond length. So, the C-C bond length is 1.1 Å, which is very close to the free (1.2 Å) and the adsorbed acetylene by calculation[2].

In the x-z planar cut at y=0.0 Å(not show here), there are two spots(A/P, B/Q) of the strongest intensity at x=±1.4 Å(z=-1.4 Å). The separation between the spot A/P and B/Q is not the actual separation between Si atom A and B, since these two spots are viewed from the two different C emitter P and Q, respectively. Instead it is the difference between the separation Si atom A and B and the C-C bond length. Then, the actual distance between them is 3.9 Å.

The spots O/Q and O/P at x=±0.55 Å(z=-2.9 Å) come from one Si atom O instead of two Si atoms, since the distance between them is relative small. Then, the distance between spots O/Q and O/P is the C-C bond length. Based on the same reasons, the actual distance between Si atom D and E is 7.5 Å instead of 6.4 Å.

According to these results, we find that the di-σ model is difficult to explain these images. The reasons are discussed in our paper[4]. If we think that acetylene adsorbs on the pedestal site, then it is easy to explain these images. That is to say, all of atomic positions given by the image are well consistent with the pedestal-site model. Some of atomic geometric parameters obtained from our image are compared in Table 1 with those from other sources. We can see that the parameters agree very well with other sources. Thus, we can conclusively think that acetylene adsorbs molecularly on the pedestal site, with no adsorption taking place on the Si dimer. Furthermore, the image provides evidence to show Si dimer is not cleaved.

Table 1. Atomic distance (Å) for Si(100)-2x1–C_2H_2 obtained from the image and other sources.

	this method	others
adsorption height	0.6	0.7[5]
C–C length	1.1	1.2
Si dimer length	2.3*	2.23[6]
Si atom A and B	3.9*	3.84[7]
Si atom D and E	7.5*	7.68[7]
$1^{st}/2^{nd}$ layer Si	0.8	1.0[6]
$2^{nd}/3^{rd}$ layer Si	1.5	1.36[7]

*C–C bond length (1.1 Å) is used. The separation between O/P and O/Q is 1.1 Å.

REFERENCES

[1] M. Nishijima, J. Yoshinobu, H. Tsuda, and M. Onchi, Surf. Sci. **192**, 383(1987).
[2] Ru-Hong Zhou, Pei-Lin Cao, and Lie-Quan Lee, Phys. Rev. B **47**, 10601(1993).
[3] Huasheng Wu and G. J. Lapeyre, Phys. Rev. B **51**, 14549(1995).
[4] S. H. Xu, Y. Yang, E. Rotenberg[1], T. Yates, Jr.[2], and G. J. Lapeyre, to be submitted.
[5] L. Li et al., Phys. Rev. B **56**, 4648(1997).
[6] N. Roberts and R. J. Needs, Surf. Sci. **236**, 112(1990).
[7] Crystal structure of bulk Si.

This work was supported by NSF. ALS of LBNL is supported by the DOE.

*email: shihong@gemini.oscs.montana.edu

Principal investigator: Gerald J. Lapeyre, Physics Department, Montana State University-Bozeman, Bozeman, MT 59717.